09/582321
PCT/FI98/01004

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E T U O I K E U S T O D I S T U S
P R I O R I T Y D O C U M E N T

Hakija Applicant	BOREALIS A/S Lyngby, Denmark
Patenttihakemus nro Patent application no	974623
Tekemispäivä Filing date	23.12.97
Kansainvälinen luokka International class	C 07F
Keksinnön nimitys Title of invention	

"A soluble magnesium dihalide complex, its preparation and use"
(Liukoinen magnesiumdihalogeenikompleksi, sen valmistus ja käyttö)

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A soluble magnesium dihalide complex, its preparation and use

The invention relates to a soluble complex comprising a magnesium dihalide and an electron donor. The invention also relates to a process for the preparation of such a complex, as well as the use of such a complex for the preparation of a polymerization catalyst component containing magnesium, transition metal, halogen and electron donor.

A complex is, according to Römpps Chemie-Lexikon, 7. Edition, Franckh'sche Verlagshandlung, W. Keller & Co., Stuttgart, 1973, page 1831, "a derived name of *compounds of higher order*, which originates from the combination of *molecules*, - unlike compounds of first order, in the creation of which atoms participate".

Generally, so called Ziegler-Natta catalyst components have been prepared by reacting a magnesium dihalide-alcohol complex compound or a magnesium alkoxide non-complex compound with a titanium halide and an electron donor which usually is a phthalic acid ester. It is necessary that the magnesium dihalide is amorphous for the catalyst component to be active. Amorphous magnesium dihalide is thus produced *in situ*.

When reacting the titanium halide with the magnesium dihalide-alcohol complex compound or the magnesium alkoxide non-complex compound, the titanium halide form titanium alkoxy trihalide, which is a harmful waste product. Both reactions have the disadvantage that the titanium halide is wasted for other purposes than the direct provision of catalytically active sites, such as chlorination of the magnesium reactant and washing away of the harmful titanium alkoxy trihalide.

In the production of amorphous magnesium dihalide, such as $MgCl_2$ strong polar ligand groups (L_1) are needed in order to break up the strong electrostatic crystallinic bonds between the $MgCl_2$ molecules according to reaction (1):

$$xMgCl_2 + xL_1 = x(Cl_2Mg\text{--}L_1) \qquad (1)$$

In practice, polar solvents are needed to carry out reaction (1). In several cases these polar solvents are reactive towards other parts of the catalyst component, and thus have to be replaced by less polar solvents (L_2). These less polar solvents are, however, often unable to react and co-ordinate with $MgCl_2$ due to the strong intermolecular forces in the $MgCl_2$ structure (2):

$$xMgCl_2 - xL_2 = \text{No reaction} \qquad (2)$$

Typical examples of solvents that are able to form complexes with $MgCl_2$ are alcohols and water. These compounds have, however, a reactive hydrogen in the hydroxyl group of their molecule which easily reacts with other compounds. Examples of less reactive solvents are the organic esters. They are less reactive towards other components but at the same time they do not have the ability to break up the strongly co-ordinated $MgCl_2$ molecules. In view of the teaching of the prior art, it seems impossible to achieve amorphous $MgCl_2$ without harmful side reactios.

The purpose of the invention is therefore to produce amorphous magnesium dihalide *in situ* without wasting titanium halide or producing harmful waste products. The invention also aims at a stoichiometric route for the preparation of Ziegler-Natta catalyst components and their intermediates. By a stoichiometric route, new catalyst components for the production of tailor-made olefin polymers can be produced.

The purposes of the invention has been achieved by means of a complex comprising a magnesium dihalide and an electron donor, which is characterized in that it is a complex of the magnesium dihalide and the electron donor and has the formula (I):

$$MgX_2 \cdot [R(OR')_n]_m \qquad (I)$$

wherein MgX_2 is the magnesium dihalide and $R(OR')_n$ is the electron donor, X is a halogen, R is an n-valent C_1-C_{20} aliphatic group, an n-valent C_7-C_{27} araliphatic group or an n-valent C_2-C_{22} acylic group, R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, n is a number from 1 to 6 and m is defined as a number $0.5 \leq m \leq 2.0$. By "n-valent acylic group" is meant a group having n acyl moieties.

Formula (I) is an empirical or semiempirical formula, i.e. m expresses the ratio between the the electron donor R(OR')n and magnesium dihalide MgX_2. The structural formula may have several molecules of MgX_2 and several same or different molecules of $R(OR')_n$, such as in the complex $(MgX_2')_a \cdot [R(OR')_n]_b$ wherein b:a = m. See e.g. formula (III) below. The claimed complex may be a statistical one, being a mixture of complexes having the average formula (I), or a specific one, essentially all the molecules of which having the same formula (I).

In the magnesium dihalide molecular component MgX_2 of the complex, X is preferably selected from Cl, Br and I, and is most preferably Cl. The most preferred complex according to the invention is thus a magnesium dichloride complex.

In the electron donor molecular component $R(OR')_n$ of the complex, R is preferably an n-valent C_2-C_{22} acylic group, preferably an n-valent aromatic C_7-C_{22} acylic group, most preferably phthaloyl. R' is preferably a C_6-C_{16} alkyl, most preferably a C_6-C_{12} alkyl like undecyl or 2-ethyl-1-hexyl. It means that the phthalate should be an ester of phthalic acid and a longer-chained alcohol. n is preferably about 1 to about 4, preferably about 2.0, meaning preference for a phthalic acid diester (phthalic acid is dibasic).

Thus, the most preferred complex is that of one of the most efficient magnesium compounds and one of the most efficient internal electron donors in the art of Ziegler-Natta catalysts, namely magnesium dichloride and a phthalic acid ester.

In the claimed complex as a whole, m depicts the ratio between the electron donor molecular component $R(OR')_n$ and the magnesium dihalide molecular component MgX_2. m is preferably about 0.67 to about 1.0, most preferably about 0.67 or about 1.0. See below, structural formulas (II) and (III).

The complex according to the invention is preferably a magnesium dichloride phthalic acid ester complex having the formula $MgCl_2 \cdot [C_6H_4(COOR')_2]_m$, wherein R' is the same as above and m is from 0.5 to 2.0, most preferably from 0.6 to 1.8.

According to one embodiment of the complex of the invention, the complex is preferably a magnesium dichloride phthalic acid ester complex having the structural formula (II):

$$MgCl_2 \cdot C_6H_4(COOR')_2 \qquad \text{(II)}$$

wherein R' is the same as above.

According to another embodiment of the complex of the invention, the complex is a magnesium dichloride phthalic acid ester complex having the structural formula (III):

$$(MgCl_2)_3 \cdot [C_6H_4(COOR')_2]_2 \qquad \text{(III)}$$

wherein R' is the same as above.

Typically, the claimed complex has an X-ray diffraction pattern (measured by a Siemens D500 instrument equipped with a Cu anode and a graphite monochromator

in the reflected beam and using an effect of 40 kV and 35 mA and a CuKα radiation wavelength of 1.541 Å), showing a dominant peak at 4.5° 2Θ.

The invention also relates to a process for the preparation of a complex comprising a magnesium dihalide and an electron donor.

Characteristic of the claimed process is that a magnesium compound (a) containing an alkoxy moiety, which magnesium compound is selected from the group consisting of a complex of a magnesium dihalide and a magnesium dialkoxide, a complex of a magnesium dihalide and an alcohol, and a non-complex magnesium dialkoxide, is reacted with a halogen compound (b), which is capable of forming the electron donor by replacement of its halogen by said alkoxy moiety.

According to the most important aspect of the invention, the process is a part of a novel stoichiometric preparation process leading to novel catalyst components for olefin polymerization.

The halogen compound (b) is capable of forming the electron donor by replacement of its halogen by said alkoxy moiety. This does not only mean that the halogen compound is a reagent leading to the electron donor, but also that it is a structural precursor thereof. Compare e.g. an alkyl halide with the corresponding dialkyl ether or an organic acid halide with the corresponding organic acid ester.

The halogen compound (b) preferably has the formula (IV):

$$RX_n \tag{IV}$$

wherein R is an n-valent C_1-C_{20} aliphatic group, an n-valent C_7-C_{27} araliphatic group or an n-valent C_2-C_{24} acylic group, X is a halogen and n is 1 to 6. In the formula, R is preferably an n-valent C_2-C_{24} acylic group, more preferably an n-valent aromatic C_7-C_{24} acylic group, most preferably phthaloyl. X is preferably selected from Cl, Br and I, and is preferably Cl. n is preferably 1 to 4, most preferably about 2.

According to a preferred embodiment of the process of the invention, said halogen compound (b) is an organic acid halide, preferably phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene. When one considers that the most preferred internal electron donor molecule of the claimed complex is a phthalic acid ester, the complexed ester molecule is simply formed by replacing the chlorines of the phthalic acid dichloride with alkoxy groups.

According to a first alternative embodiment of the process of the invention, said magnesium compound (a) containing an alkoxy moiety is a magnesium dichloride-magnesium dialkoxide complex of the formula (V):

$$MgCl_2 \cdot [Mg(OR')_2]_p \qquad (V)$$

wherein R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group, and p is 1 to 6, preferably about 2. Formula (V) is empirical or semiempirical, meaning that the complex's molecular clusters can consist of several $MgCl_2$ molecules and several $Mg(OR')_2$ molecules, corresponding to the formula $(MgCl_2)_c[Mg(OR')_2]_d$ wherein d:c = p. In the process of the invention, said magnesium dichloride-magnesium dialkoxide complex is reacted with the above halogen compound so that the halogen compound (b) has its halogen(s) replaced by the alkoxide(s) of the complex and forms an electron donor, whereby the complex of magnesium dichloride and the electron donor is formed.

Preferably, said complex of a magnesium dihalide and a magnesium dialkoxide is a magnesium dichloride-dimagnesium dialkoxide complex of the structural formula (VI):

$$MgCl_2 \cdot [Mg(OR')_2]_2 \qquad (VI)$$

wherein R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group.

Said magnesium dichloride-magnesium dialkoxide complex is preferably prepared by reacting magnesium dichloride and an alcohol into an intermediate which is a magnesium dichloride-alcohol complex $MgCl_2 \cdot (R'OH)_{2p}$ wherein R' is the same as above, and reacting the magnesium dichloride alcohol complex with p mol of a magnesium dialkyl MgR''_2, wherein R'' is a hydrocarbyl group having 1 to 20 carbon atoms. If R'' is a hydrocarbyl group having 1 to 5 carbon atoms, a volatile alkane R''H byproduct is thereby formed and easily removed by evaporation. In the synthesis, the molar ratio $MgCl_2$:R'OH is preferably between 1:1 and 1:8, most preferably between 1:2 and 1:5. The molar ratio $MgCl_2 \cdot (R'OH)_{2p}:MgR''_2$ is preferably between 1:1 and 1:4, most preferably about 1:2.

Said magnesium compound (a) which is said magnesium dichloride-dimagnesium dialkoxide complex $MgCl_2 \cdot [Mg(OR')_2]_2$, wherein R' is the same as above, is preferably reacted with said halogen compound (b) which is said phthalic acid

dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene. Typically, the product is $(MgCl_2)_3 \cdot [Ph(COOR')]_2$. See formula (III) above.

In the first alternative embodiment of the claimed process, said magnesium compound (a) and said halogen compound (b) are reacted in essentially stoichiometrical amounts and, independently, at a temperature of between 80 °C and 160 °C. The reaction time is preferably about 2h to about 8 h.

Most preferably, the magnesium dihalide and the alcohol, which is a heavier alcohol, are first reacted at a temperature between 120 °C and 160 °C, after which the product is reacted with the magnesium alkoxide at a temperature between 80 °C and 120 °C, followed by reaction with the halogen compound at said lower temperature interval.

A typical example of said first alternative embodiment of the claimed process is described in figure 1.

According to a second alternative embodiment of the claimed process, said magnesium compound (a) containing an alkoxy moiety is a non-complex magnesium dialkoxide of the formula (VII):

$$Mg(OR')_2 \qquad \text{(VII)}$$

wherein R' is a C_1-C_{20} aralkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group.

Said non-complex magnesium dialkoxide is preferably prepared by reacting a magnesium dialkyl, preferably a magnesium dialkyl of the formula MgR''_2, wherein R" is a hydrocarbyl group having 1 to 20 carbon atoms, and an alcohol, preferably an alcohol of the formula R'OH, wherein R' is the same as above. The preferred ratio between the magnesium dialkyl and the alcohol is about 1:2.

In the second alternative embodiment of the claimed process, said magnesium compound (a) which is said non-complex magnesium dialkoxide has the formula $Mg(OR')_2$, wherein R' is a C_1-C_{20} aralkyl or a C_7-C_{27} aralkyl, preferably a C_6-C_{16} alkyl, is preferably reacted with said halogen compound (b) which is said phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene. Usually, said magnesium compound (a) and said halogen compound (b) are reacted in essentially stoichiometric amounts.

A typical example of said second alternative embodiment of the claimed process is described in figure 2.

According to a third alternative embodiment of the process of the invention, said magnesium compound (a) containing an alkoxy moiety is a complex of a magnesium dihalide and an alcohol of the formula (VIII):

$$MgCl_2 \cdot (R'OH)_q \qquad \text{(VIII)}$$

wherein R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group, and q is from 1 and 6. This complex is often used as starting material for Ziegler-Natta catalyst components. However, it is not known to have been used as starting material for a magnesium dihalide electron donor complex by reacting it with a halogenous electron donor precursor in the above described way.

The complex of a magnesium dihalide and an alcohol is usually prepared by reacting magnesium dichloride $MgCl_2$ and and alcohol R'OH, wherein R' is the same as above.

In the process according to the second alternative embodiment, said magnesium compound (a) which is said complex of a magnesium dihalide and an alcohol having the formula $MgCl_2 \cdot (R'OH)_q$, wherein R' is a C_1-C_{20} alkyl or a C_7-C_{27} aralkyl, preferably a C_6-C_{16} alkyl, and q is from 1 and 6, is reacted with said halogen compound (b) which is said phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-~~phenylene. An example of this third alternative embodiment is described in figure 3.~~

Above, the product and its preparation according to the invention have been described. As the claimed complex finds its natural application in the field of olefin polymerization catalyst synthesis, the invention also relates to the use of said complex in that field. Thus, claimed is the use of said complex for the preparation of a polymerization catalyst component containing magnesium, as well as at least one transition metal, halogen and electron donor. More specifically, the use is characterized in that said complex is reacted with a titanium halide (c) to give said catalyst component.

Said titanium halide (c) preferably has the formula (IX):

$$(OR''')_p TiX_{4-p} \qquad \text{(IX)}$$

wherein R''' is a C_1-C_{10} alkyl group or a C_7-C_{16} aralkyl group, X is a halogen and p is 0 to 3. Most preferably, said titanium halide (c) is a titanium tetrahalide TiX_4, wherein X is the same as above, most preferably titanium tetrachloride $TiCl_4$.

Experimental

Preparation of the complexes

All chemicals were handled in strict inert conditions and all the reactions took place also in strict inert conditions in a nitrogen atmosphere.

Example 1 (first alternative embodiment)

First 1.068 g (11.2 mmol) of $MgCl_2$ was introduced into a 50 ml glass reactor. 9.60 ml (8.0 g, 46.2 mmol) of 1-undecanol was added on to the $MgCl_2$. The slurry was mixed using a magnetic stirring bar and the solution was heated to 130 °C and the reactants were allowed to react with each other at this temperature for 3 h. The slurry was cooled down to 100 °C and 9.6 ml (8.3 g, 90 mmol) of toluene was added to the reaction solution to increase its dissolving capability. 25.40 ml (18.52 g, 22.3 mmol R''_2Mg) of a 20% heptane solution of butyl-octyl magnesium was now introduced. Finally, 3.24 ml (4.565 g, 22.5 mmol) of phthaloyl chloride was added. The achieved product was dried under a stream of nitrogen for several hours at temperatures between 90 °C and 120 °C.

Example 2 (second alternative embodiment)

123.2 mmol of butyl-octyl magnesium was introduced into a 250 ml glass reactor. A 20% heptane solution of the butyl-octyl magnesium containing 2.92 w-% of Mg was used giving a feed volume of 139.4 ml (102.5 g) in the reactor. 244.6 mmol (38.2 ml, 31.85 g) of 2-ethyl-1-hexanol was then added slowly at room temperature. The addition of alcohol took 23 min. Mixing speed was 242 rpm. The temperature was increased to 63 °C and the reactants were allowed to react with each other at that temperature for 15 min. After this 122.74 mmol (17.69 ml, 24.92 g) of phthaloyl chloride was added slowly at room temperature. The temperature was increased during 10 min to 50 °C and the reactants were again allowed to react with each other at that temperature for 5 min. After this the reaction solution was allowed to cool down to room temperature.

28.8 g of the achieved solution was taken into a 100 ml glass reactor for solvent evaporation. The sample was dried under vacuum in a stream of nitrogen gas at 50 °C for 3 h. 12 ml of condensed solvent (heptane) was trapped in the vacuum trap.

The product was washed with 60 ml of pentane at 45 °C for 45 min, after which the product was allowed to settle for 45 min and the solid product was separated from the solution. The product was washed a second time with 44 ml of pentane and finally dried under vacuum and in a stream of nitrogen at 50 °C for one hour.

Example 3 (third alternative embodiment)

22.5972 mmol (2.152 g) of $MgCl_2$ was introduced into a 100 ml glass reactor. To this, 45.193 mmol (7.101 ml, 5.915 g) of 2-ethyl-1-hexanol was added. Finally, 22.5972 mmol (3.256 ml, 4.588 g) of phthaloyl dichloride was added to the mixture. The mixture was kept under agitation at 60 °C for 30 min. A solid complex was collected from the vessel by evaporation and washed three times with a 100 ml portion of heptane at 90 °C for 15 min, then with a 100 ml portion of pentane at room temperature and finally dried. This product was reacted with $TiCl_4$ into a catalytically active complex.

Comparative example

A fourth sample was prepared by introducing 20 mmol $MgCl_2$ (1.90422 g) in a 150 ml glass reactor equipped with a magnetic stirrer. 20 mmol of di-2-ethyl-hexyl-phthalate (8.0 ml, 7.81 g) (DOP) was then added on to the $MgCl_2$. The reactants were allowed to react with each other overnight. The product was washed with pentane and dried in the same way as described above.

Characterization of the products by x-ray diffractometry and Infrared spectroscopy

The products were characterized by infrared spectroscopy (IR) and by taking X-ray diffraction patterns of it. The WAXS patterns were collected in reflection mode between 2 and 70°2 θ with a Siemens D500 instrument. The diffractometer was equipped with a Cu anode and a graphite monochromator in the reflected beam. The effect used was 40 kV and 35 mA. The CuKα radiation wavelength was 1.541 Å. The sample was loaded in a glove box into a Mylar film covered sample holder. The Mylar film forms a half-cylindrical window, allowing X-rays to pass perpendicularly through.

Di-undecylphthalate (DUP) was used as electron donor reference in the IR studies. The products of examples 1 and 2 were investigated together with the standard DUP. The products of example 1 was analyzed twice: right away and a second time after overnight storage.

The IR spectra were taken by a Nicolet 510 FTIR equipment with 2 cm^{-1} resolution. The number of scans were 128. All the samples were investigated as capillary films between two KBr pellets. Pure DUP was not handled in inert conditions, but the $MgCl_2$ samples were handled in a glove box in an inert nitrogen environment in order to protect the samples from air and moisture. To get thin enough capillary films the samples were somewhat heated when placed in between the KBr pellets.

X-ray characterization

The X-ray diffraction pattern of the $MgCl_2$-DOP complex is shown in figure 4. The pattern show no sign of $MgCl_2$. At 50 ° 2Θ, where the most significant peak of pure $MgCl_2$ is to be found, there is no sign of a peak. This is also the case at 30° and 35° 2Θ. On the other hand, there is a new dominant peak at 4.5° 2Θ showing that the reflecting layers have been separated far from each other (21 Å).

The X-ray diffraction pattern of the product coming from the direct reaction between $MgCl_2$ and DOP of the comparative example is shown in figure 5. The pattern showed that no reaction between these components had occurred, the X-ray pattern showed pure $MgCl_2$ with its significant reflecting signals at 15°, 30°, 35° and 50° 2Θ.

The results show that in the first three synthesis routes the donor compound had been complexed with $MgCl_2$ at a molecular level, thereby breaking up the strong molecular structure of crystalline $MgCl_2$ and indicating in situ preparation thereof. The results show also that in the fourth, direct synthesis route, the donor compound had not been able to form a complex with $MgCl_2$ but the product contained only original $MgCl_2$.

Results by Infrared Spectroscopy

The samples were prepared and studied by IR spectroscopy according to the description in the experimental section above. As $MgCl_2$ does not have any absorbance spectrum in the IR area from 4000 to 400 cm^{-1}, the IR study concentrated on the changes in the structure of the DOP and DUP complexation to the $MgCl_2$.

The pure DOP and DUP absorbs IR light due to the stretching vibration of the ester carbonyl double bond of the C=O group at the wavelength of 1729 cm^{-1}. The corresponding stretching vibration of the C-O- bond is to be found at 1280 cm^{-1} and at 1100 cm^{-1} (see figure 6).

When the $MgCl_2$ was co-ordinated to DOP in the molar ratio of 1:1, clear changes in the IR spectrum of DOP could be seen (figure 6, Example 2). The absorption peaks of pure DOP could still clearly be seen which indicates that a part of the carbonyl groups are still unco-ordinated. On the right side of the original C=O absorption peak there is a new shoulder indicating that a part of the C=O groups have co-ordinated to Mg causing the double bond of the C=O group to loosen up starting to resemble more a single C-O bond. The proportionally small shift in the position of the "shoulder peak" indicate a weak interaction, i.e. a weak co-ordination of $MgCl_2$ to the C=O oxygen. A weak interaction is also indicated by the sign of several secondary "shoulder" peaks.

When the $MgCl_2$ amount was increased by 50% in the complex (Example 1), the proportion of the "shoulder" peak at about 1690 cm^{-1} increased. There is however no sign of a strong co-ordination (figure 6). The changes in the IR spectra show up even better in the sample that had been stored overnight. Here the main peak of the carbonyl oxygen has shifted from the position of 1729 cm^{-1} for the pure DUP to 1719 cm^{-1}. The results show also that the co-ordinated carboxyl group is influencing the "free" carboxyl group as the position of its peak is shifted 10 cm^{-1}.

The same results can be seen in connection with the absorption peak of the C-O- bond. The absorption peak of the C-O- bond in the pure DUP is found at 1287 cm^{-1}. Looking at the spectra for the $MgCl_2 \cdot DOP$ (Example 2) and the $(MgCl_2)_{1.5} \cdot DUP$ (Example 1) samples there is a corresponding "shoulder" peak forming to the left of the original peak indicating a weak double bond character of the C-O bond. This shift is so strong that the original peak at 1287^{-1} is not any longer detectable in the product that had been stored overnight (figure 6). These results indicate that the Mg in the $MgCl_2$ is complexed between the C=O oxygen and the C-O oxygen atoms in the $MgCl_2 \cdot DOP$ and in the $MgCl_2 \cdot DUP$ complex.

Conclusively it can be said that in the IR spectrum of the pure DUP the peaks show unco-ordinated carbonyl groups, in the $MgCl_2 \cdot DOP$ and in the MgCl2·DUP sample (Example 2) there is a $MgCl_2$ co-ordination to one of the carbonyl groups, the other being free, and in the $(MgCl_2)_{1.5} \cdot DUP$ sample (Example 1) there is a partial co-ordination of $MgCl_2$ to both of the carbonyl groups.

Summary

In this study a stoichiometric synthesis route was used to produce the $MgCl_2$-$C_6H_4(COOR')_2$ complex. $MgCl_2$ and Mg-alkyl have been reacted with an alcohol to form a $MgCl_2$-Mg-alcoholate complex, a Mg-alcoholate and an $MgCl_2$-alcohol complex. These Mg-alcoholates or $MgCl_2$-alcohol complex has then been brought into contact with phthaloyl chloride to give an $MgCl_2$-donor complex. It was not possible to produce an $MgCl_2$-donor complex of this type through a direct contact between $MgCl_2$ and the corresponding donor, which rules out the possibility that this complex could have been formed unintactionally in the prior art. The complex achieved trough the synthesis routes described in this study is identifiable through its distinct X-ray diffraction pattern that shows a dominant peak at 4.5° 2Θ. IR studies showed that the Mg in the $MgCl_2$ is co-ordinated both to the C=O oxygen and the C-O- oxygen in the ester group.

Claims

1. A complex comprising a magnesium dihalide and an electron donor, **characterized** in that it is a complex of the magnesium dihalide and the electron donor and has the formula (I):

$$MgX_2 \cdot [R(OR')_n]_m \qquad (I)$$

wherein MgX_2 is the magnesium dihalide and $R(OR')_n$ is the electron donor, X is a halogen, R is an n-valent C_1-C_{20} aliphatic group, an n-valent C_7-C_{27} araliphatic group or an n-valent C_2-C_{22} acylic group, R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, n is a number from 1 to 6 and m is defined as a number $0.5 \leq m \leq 2.0$.

2. The complex according to Claim 1, **characterized** in that X is selected from Cl, Br and I, and is preferably Cl.

3. The complex according to Claim 1 or 2, **characterized** in that R is an n-valent C_2-C_{22} acylic group, preferably an n-valent aromatic C_7-C_{22} acylic group, most preferably phthaloyl.

4. The complex according to Claim 1, 2 or 3, **characterized** in that R' is a C_6-C_{16} alkyl group, preferably a C_6-C_{12} alkyl group like undecyl or 2-ethyl-1-hexyl.

5. The complex according to any preceding claim, **characterized** in that n is about 1 to about 4, preferably about 2.0.

6. The complex according to any preceding claim, **characterized** in that m is about 0.67 to about 1.0.

7. The complex according to any preceding claim, **characterized** in that it is a magnesium dichloride phthalic acid ester complex having the formula (II):

$$MgCl_2 \cdot C_6H_4(COOR')_2 \qquad (II)$$

wherein R' is the same as above.

8. The complex according to one of Claims 1 to 5, **characterized** in that it is a magnesium dichloride phthalic acid ester complex having the formula (III):

$$(MgCl_2) \cdot [C_6H_4(COOR')_2]_2 \qquad (III)$$

wherein R' is the same as above.

9. The complex according to any preceding claim, **characterized** in that it has an X-ray diffraction pattern showing a dominant peak at 4.5°2Θ.

10. Process for the preparation of a complex comprising a magnesium dihalide and an electron donor, **characterized** by reacting a magnesium compound (a) containing an alkoxy moiety, which magnesium compound is selected from the group consisting of a complex of a magnesium dihalide and a magnesium dialkoxide, a complex of a magnesium dihalide and an alcohol, and a non-complex magnesium dialkoxide, with a halogen compound (b), which is capable of forming the electron donor by replacement of its halogen by said alkoxy moiety.

11. Process according to Claim 10, **characterized** in that said halogen compound (b) has the formula (IV):

$$RXn \qquad \text{(IV)}$$

wherein R is an n-valent C_1-C_{20} aliphatic group, an n-valent C_7-C_{27} araliphatic group or an n-valent C_2-C_{24} acylic group, X is a halogen and n is 1 to 6.

12. The complex according to Claim 11, **characterized** in that R is an n-valent C_2-C_{24} acylic group, preferably an n-valent aromatic C_7-C_{24} acylic group, most preferably phthaloyl.

13. Process according to Claim 11 or 12, **characterized** in that X is selected from Cl, Br and I, and is preferably Cl.

14. Process according to Claim 11, 12 or 13, **characterized** in that n is 1 to 4, preferably about 2.

15. Process according to any of Claims 11 to 14, **characterized** in that said halogen compound is an organic acid halide, preferably phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene.

16. Process according to any of Claims 10 to 15, **characterized** in that said complex of a magnesium dihalide and a magnesium dialkoxide is a magnesium dichloride-magnesium dialkoxide complex of the formula (V):

$$MgCl_2 \cdot [Mg(OR')_2]_p \qquad \text{(V)}$$

wherein R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group, and p is 1 to 6, preferably about 2.

17. Process according to Claim 16, **characterized** in that said complex of a magnesium dihalide and a magnesium dialkoxide is a magnesium dichloride-dimagnesium dialkoxide complex of the formula (VI):

$$MgCl_2 \cdot [Mg(OR')_2]_2 \qquad \text{(VI)}$$

wherein R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group.

18. Process according to Claim 16, **characterized** in that said magnesium dichloride magnesium dialkoxide complex is prepared by reacting magnesium dichloride with and alcohol into an intermediate which is a magnesium dichloride alcohol complex $MgCl_2 \cdot (R'OH)_{2p}$, wherein R' is the same as above, and reacting the magnesium dichloride alcohol complex with p mol of a magnesium dialkyl MgR''_2, wherein R" is a hydrocarbyl group having 1 to 20 carbon atoms.

19. Process according to Claim 18, **characterized** in that, independently, the molar ratio $MgCl_2$:R'OH is between 1:1 and 1:8, preferably between 1:2 and 1:5, the molar ratio $MgCl_2 \cdot (R'OH)_{2p}:MgR''_2$ is between 1:1 and 1:4, preferably about 1:2, the temperature is between 80 °C and 160 °C, and the reaction time is about 2 h to about 8 h.

20. Process according to Claim 15 and 17, **characterized** in that said magnesium compound (a) which is said magnesium dichloride-dimagnesium dialkoxide complex $MgCl_2 \cdot [Mg(OR')_2]_2$, wherein R' is a C_6-C_{16} alkyl group, is reacted with said halogen compound (b) which is said phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene.

21. Process according to any of Claims 10 to 15, **characterized** in that said non-complex magnesium dialkoxide has the formula (VIII):

$$Mg(OR')_2 \qquad \text{(VIII)}$$

wherein R' is a C_1-C_{20} aralkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group.

22. Process according to Claim 21, **characterized** in that said non-complex magnesium dialkoxide is prepared by reacting a magnesium dialkyl, preferably a

magnesium dialkyl of the formula MgR''_2, wherein R" is a hydrocarbyl group having 1 to 20 carbon atoms, and an alcohol, preferably an alcohol of the formula R'OH wherein R' is the same as above.

23. Process according to Claim 15 and 21, **characterized** in that said magnesium compound (a) which is said non-complex magnesium dialkoxide has the formula $Mg(OR')_2$, wherein R' is a C_1-C_{20} aralkyl or a C_7-C_{27} aralkyl, preferably a C_6-C_{16} alkyl, is reacted with said halogen compound (b) which is said phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene.

24. Process according to any of Claims 10 to 15, **characterized** in that said complex of a magnesium dihalide and a magnesium dialkoxide is a complex of a magnesium dihalide and an alcohol having the formula (VII):

$$MgCl_2 \cdot (R'OH)_q \qquad \text{(VII)}$$

wherein R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, preferably a C_6-C_{16} alkyl group, and q is from 1 to 6.

25. Process according to Claim 24, **characterized** in that said complex of a magnesium dihalide and an alcohol is prepared by reacting magnesium dichloride $MgCl_2$ and and alcohol R'OH, wherein R' is the same as above.

26. Process according to Claim 24 or 25, **characterized** in that the reaction temperature is kept between 10 °C and 100 °C, and the reaction time is about from 10 to about 90 min.

27. Process according to Claim 15 and 24, **characterized** in that said magnesium compound (a) which is said complex of a magnesium dihalide and an alcohol having the formula $MgCl_2 \cdot (R'OH)_q$, wherein R' is a C_1-C_{20} alkyl or a C_7-C_{27} aralkyl, preferably a C_6-C_{16} alkyl, and q is from 1 to 6, is reacted with said halogen compound (b) which is said phthalic acid dichloride $Ph(COCl)_2$, wherein Ph is o-phenylene.

28. Process according to one of Claims 10 to 27, **characterized** in that said magnesium compound (a) and said halogen compound (b) are reacted essentially stoichiometrically.

29. Use of a complex according to one of Claims 1 to 9 or a complex prepared according to one of Claims 10 to 28 for the preparation of a polymerization catalyst component containing magnesium, transition metal, halogen and electron donor.

30. Use according to Claim 29, **characterized** in that said complex is reacted with a titanium halide (c).

31. Use according to Claim 30, **characterized** in that said titanium halide (c) has the formula (IX):

$$(OR'''')_pTiX_{4-p} \qquad (IX)$$

wherein R"" is a C_1-C_{10} alkyl group or a C_7-C_{16} aralkyl group, X is a halogen and p is 0 to 3, and preferably is a titanium tetrahalide TiX_4, wherein X is the same as above, most preferably titanium tetrachloride $TiCl_4$.

32. A complex according to one of claims 1 to 31, **characterized** in that it shows an IR spectra with an absorption peak for the C=O...Mg that has shifted 5 to 15 cm^{-1}, preferably 10 cm^{-1} to the right, and preferably also shows three shoulders.

(57) **Abstract**

The invention relates to a complex comprising a magnesium dihalide and an electron donor. It is a complex of the magnesium dihalide and the electron donor and has the formula (I):

$$MgX_2 \cdot [R(OR')_n]_m \qquad \text{(I)}$$

wherein MgX_2 is the magnesium dihalide and $R(OR')_n$ is the electron donor, X is a halogen, R is an n-valent C_1-C_{20} aliphatic group, an n-valent C_7-C_{27} araliphatic group or an n-valent C_2-C_{22} acylic group, R' is a C_1-C_{20} alkyl group or a C_7-C_{27} aralkyl group, n is a number from 1 to 6 and m is defined as a number $0.5 \leq m \leq 2.0$. The invention also relates to the preparation process of such a complex, as well as the use thereof for the preparation of olefin polymerization catalyst components.

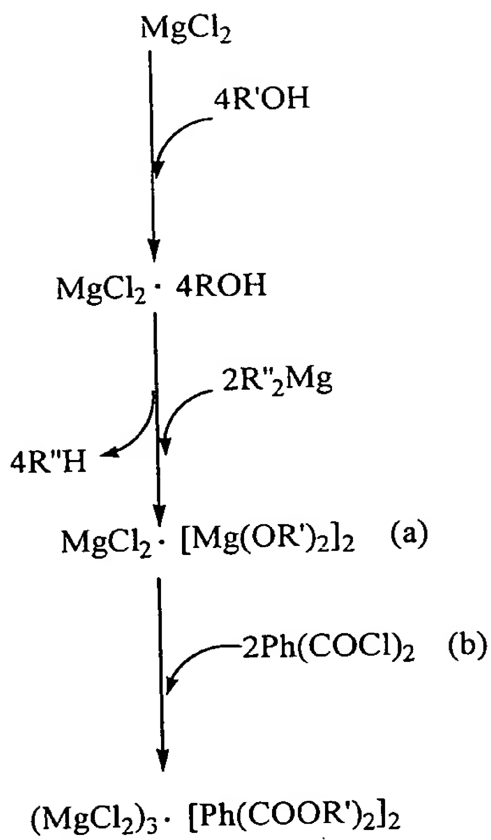

Figure 1 Example of first alternative embodiment

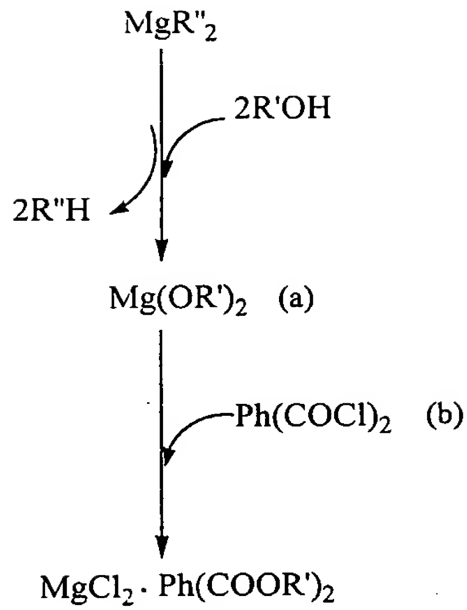

Figure 2 Example of second alternative embodiment

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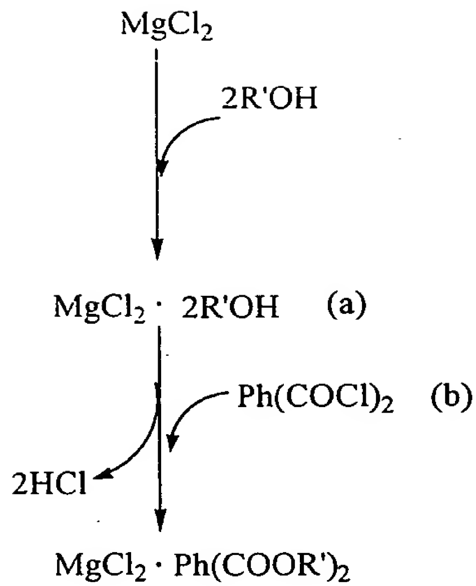

Figure 3 Example of third alternative embodiment

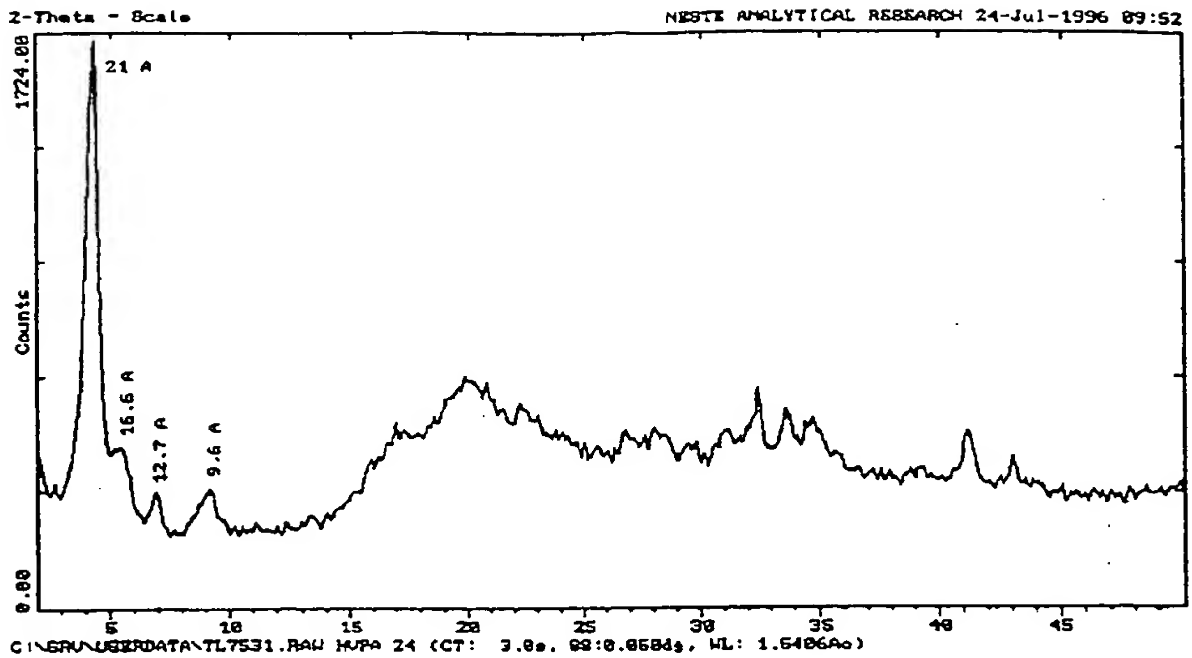

Figure 4 The x-ray diffraction pattern of the Mg-complex produced out of Mg-alkyl, alcohol and phthalic ester (example 2)

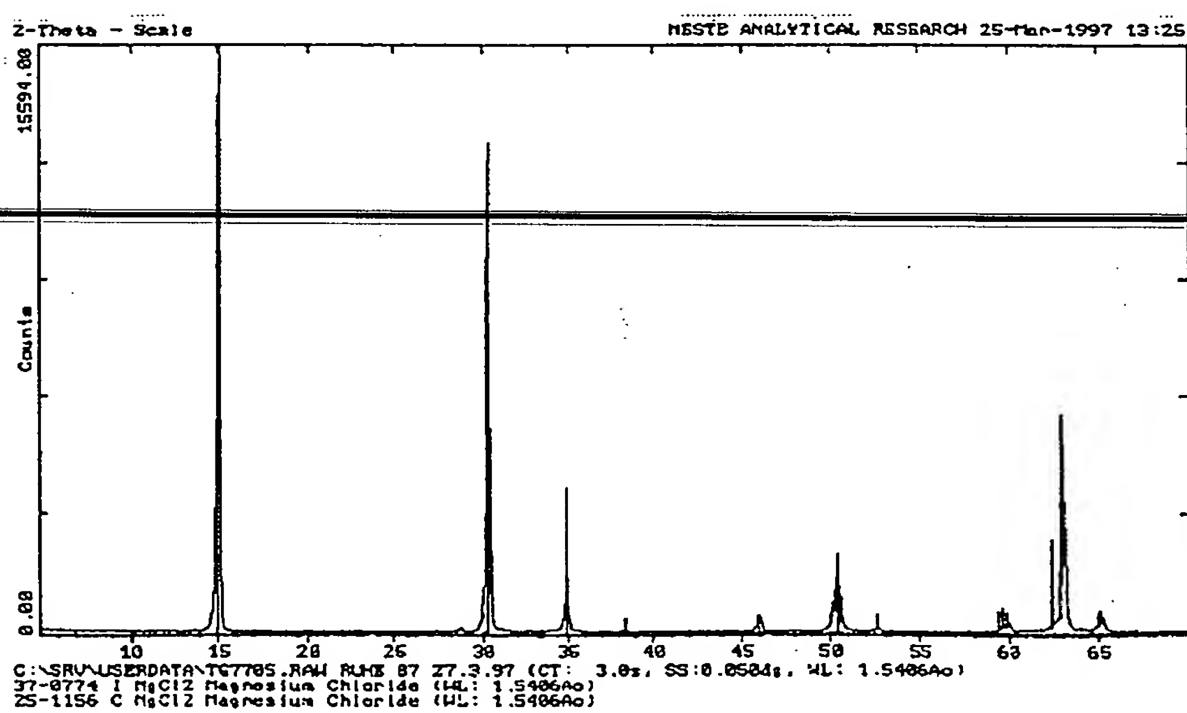

Figure 5 The x-ray diffraction pattern of the product of $MgCl_2$ and phthalic ester (comparative example)

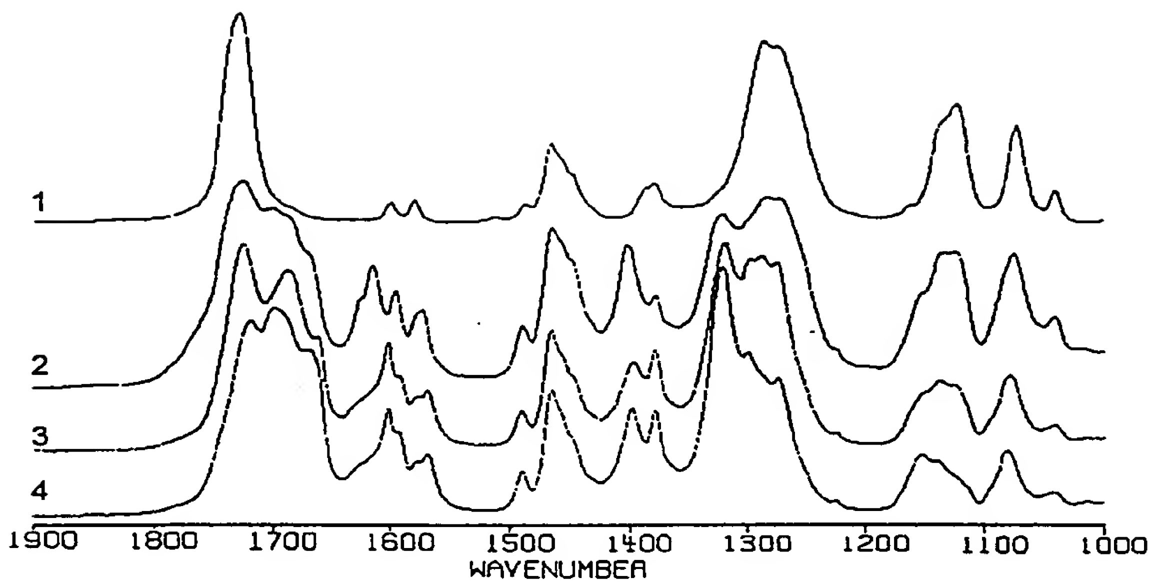

Figure 6 IR Spectra of

1 DUP,

2 $MgCl_2 \cdot$ DOP (example 2),

3 $(MgCl_2)_{1.5} \cdot$ DUP (example 1) and

4 $(MgCl_2)_{1.5} \cdot$ DUP (example 1) stored overnight